

# Magneto-Optical Kerr Effect of Iron Thin Films on Paramagnetic Substrates

A. Debernardi,<sup>a,b</sup> I. Galanakis,<sup>c</sup> M. Alouani,<sup>a,\*</sup> and H. Dreyssé<sup>a</sup>

<sup>a</sup> *Institut de Physique et Chimie des matériaux de Strasbourg,  
UMR 7504 du CNRS, Université Louis Pasteur, 23,  
rue du Loess, 67037 Strasbourg, France, European Union*

<sup>b</sup> *Istituto Nazionale per la Fisica della Materia (INFM),  
Udr Trieste Università/SISSA, Dip. Fisica Teorica,  
Strada Costiera 11, 34014 - Trieste, Italy, European Union*

<sup>c</sup> *Institut für Festkörperforschung, Forschungszentrum Jülich, D-52425 Jülich, Germany, European Union*  
(Dated: February 1, 2008)

First principles calculations of the magnetic properties and the magneto-optical Kerr effect (MOKE) of iron thin films epitaxially grown on the [001] surface of paramagnetic metals: copper, silver, gold, palladium, and platinum are presented. The role of hybridization with the substrate is investigated and it is shown how the relaxation effects influence the complex Kerr angle. The results are obtained by means of the relativistic full-potential linear muffin-tin method, and the film is modeled using a slab geometry within a supercell technique.

PACS numbers: 71.15, 75.70, 78.20Ls

The magneto-optical Kerr effect (MOKE) has become a standard technique for studying the magnetic properties of a variety of low dimensional systems like films, surfaces or multilayers<sup>1</sup>. This effect occurs when plane polarized light is reflected by a magnetic material; the light becomes elliptically polarized and the plane of polarization rotates. Typically, this effect is not very large for the 3d materials but it yields important information for their magnetic properties as it is sensitive to the local environment of the 3d atoms. Although a theoretical investigation has already been performed to study the effect of the lattice parameter on the MOKE of bulk 3d metals,<sup>2</sup> it failed to explain the differences in MOKE seen for thin films grown on different substrates. In this contribution we use the relativistic full-potential linear muffin-tin method<sup>3</sup> within the local-spin density approximation<sup>4</sup> to calculate the complex Kerr angle and the magnetic moments of an iron monolayer epitaxially grown on the (001) surface of five different paramagnetic substrates: copper, palladium, platinum, gold, and silver. These materials, that belong to two adjacent column of the periodic table, have a face centered cubic structure and - due to similar electronic properties and lattice parameter - allow to study the trends of magnetic properties of Fe-layer as a function of orbital hybridization and of the lattice constant of the substrate. We show that the substrate influences the MOKE in a more dramatic way than the one suggested just by the change of the Fe lattice parameter.

We consider elliptically polarized light and we define as  $\epsilon_K$  the ellipticity and as  $\theta_K$  the rotation angle of the major axis compared to the incident beam. In the case that the magnetization axis is perpendicular to the surface the in-plane elements of the conductivity tensor are decoupled from the perpendicular orientation and

$\sigma_{xx} = \sigma_{yy}$ ;  $\sigma_{xy} = -\sigma_{yx}$ . If we consider also the case of the polar Kerr effect, i.e. the incident beam is perpendicular to the plane, then the expression of the complex Kerr angle  $\theta_K + i\epsilon_K$  becomes

$$\theta_K + i\epsilon_K = \frac{-\sigma_{xy}}{\sigma_{xx} \sqrt{1 + \frac{4\pi i}{\omega} \sigma_{xx}}} \quad (1)$$

for small Kerr angles as is the case for the transition metals. The implementation of the calculation of the conductivity tensor in our electronic structure method is described in Ref. 2. In our calculations we do not take into account intraband transitions, which are usually described by the phenomenological Drude model, but this will affect only the spectra below 0.5 eV and thus will not change the following discussion.

The dielectric tensor  $\epsilon$  is related to the complex conduction tensor  $\sigma \equiv \sigma^{(1)} + i\sigma^{(2)}$  (as usual,  $\sigma^{(1)}$  and  $\sigma^{(2)}$  denote respectively the real part and the coefficient of the imaginary part) through the simple relation

$$\epsilon = 1 + \frac{4\pi i}{\omega} \sigma. \quad (2)$$

Thus if the complex Kerr angle is known from experiment, one can extract the dielectric function of a material.

In our calculation we use a slab structure within a super-cell geometry. The slab is composed of five layers of paramagnetic material, with a monolayer of iron at each extremity of the paramagnetic substrate, so that no slab-dipole is created. We found that five substrate layers are sufficient so that the central layer of Cu (Pd, Pt, Au, or Ag) is bulklike. In all these calculations we used the experimental bulk lattice parameters for the substrate and the plane perpendicular to the grown direction (the values are displayed in Tab. I). We also converged the vacuum between two adjacent slabs and we found that a vacuum of three times the bulk lattice parameter of the substrate metal is sufficient so that no

---

\*Present Address: KAVLI Institute for Theoretical Physics, Santa Barbara, Ca 93106-4030, USA

TABLE I: Calculated spin and orbital magnetic moments in  $\mu_B$  for both the Fe overlayer and the first noble-metal sub-surface layer for all three noble metals. We also present the experimental lattice bulk parameters  $a_{bulk}$  of the substrate metals and the interlayer distance  $d$  between the Fe layer and the substrate in units of the substrate lattice constant.

System	$a_{bulk}$ (Å)	$d$ ( $a_{bulk}$ )	$m_{Fe}^{spin}$ ( $\mu_B$ )	$m_{Fe}^{orb}$ ( $\mu_B$ )	$m_{Sub}^{spin}$ ( $\mu_B$ )	$m_{Sub}^{orb}$ ( $\mu_B$ )
Fe/Cu(001)	3.61	0.4703	2.728	.070	.055	.008
Fe/Pd(001)	3.89	0.4228	3.053	.090	.340	.025
Fe/Pt(001)	3.92	0.4200	2.982	.075	.308	.040
Fe/Au(001)	4.08	0.4005	2.978	.070	.062	.022
Fe/Ag(001)	4.09	0.4086	2.981	.107	.013	.004

inter-slab interactions occur.<sup>5</sup> Since the first iron monolayer on the (001) surface grows pseudomorphically, we have considered only the relaxation of the Fe layer with respect to the rigid substrate. The equilibrium geometry is obtained by a polynomial fit of the total energies computed for different interlayer distances.<sup>6</sup> We found that the relaxation of iron layer is essential to properly compute magneto-optical effects.<sup>6</sup> Our results of Fe orbital and spin magnetic moments together with the first layer substrate induced spin magnetic moments and orbital moments are reported in Tab. I. For the materials under study the largest contractions of the Fe layer along the growth axis are found for Ag and Au substrates, to compensate for the larger in-plane expansion of Fe in the Ag-(Au)-based system compared to the other ones. Note that along the [001] direction the distance between two successive layers for an fcc structure is 0.5 times the bulk lattice parameter; as the lattice parameter of the substrate becomes closer to that of the bulk iron the interlayer distance  $d$  approaches the “ideal” value of 0.5.

The Kerr rotation angle and the ellipticity in the polar configuration in the case of Fe layer grown on (001) surface of Cu, Pd, Pt, Au, and Ag<sup>7</sup> substrates are displayed in Figs 1, and 2 respectively. For completeness and for further discussion in Fig. 3 we have reported the computed real and imaginary part of in-plane conductivity tensor.

For Cu substrate a small enhancement of the Kerr angle  $\theta_K$  occurs at  $\sim 2.21$  eV which corresponds to the plasma edge of the bulk copper metal.<sup>8</sup> The strong enhancement at an energy of  $\sim 5.5$  eV and  $\sim 6.5$  eV for  $\theta_K$  and  $\epsilon_K$ , respectively, corresponds to the depletion detected already in bulk iron. Similar feature are also displayed in Fig. 1 for Au substrate where the signal is magnified according to the fact that in this system the magnetization of iron and of the top substrate layer are larger than for Fe/Cu(001) (see Tab. I). Our results for the magneto-optical Kerr effect for Fe/Ag(001) are presented in Fig. 1. The richness of structures displayed in the relaxed spectra of Fe/Cu(001) or Fe/Au(001) is absent in our computed Kerr angles for Fe/Ag(001) since,

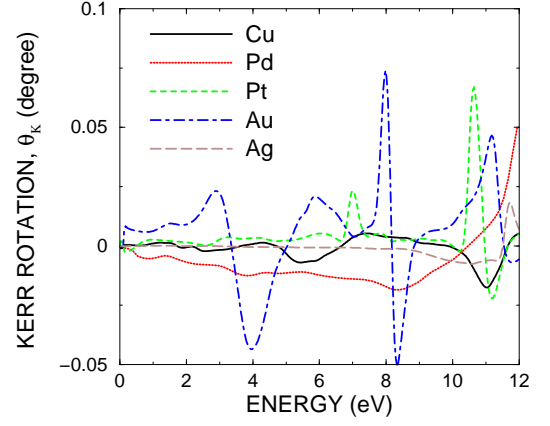


FIG. 1: Magneto-optical Kerr rotation angle for Fe/Cu(001), Fe/Pd(001), Fe/Pt(001), Fe/Au(001) and Fe/Ag(001) systems in the polar configuration.

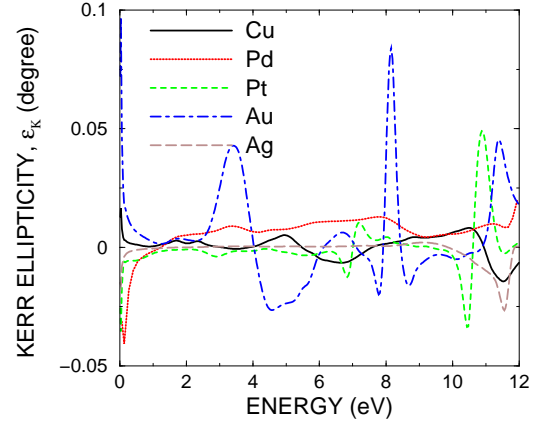


FIG. 2: Magneto-optical Kerr ellipticity for Fe/Cu(001), Fe/Pd(001), Fe/Pt(001), Fe/Au(001) and Fe/Ag(001) systems in the polar configuration.

in the latter system, they became significant only for energies higher than  $\sim 10$  eV. This can be explained by observing the conductivity tensor of this system is smooth as a function of energy. Note that the first Ag-layer display the lower magnetization among all the substrate under investigation.

In spite that Fe/Pd(001) and Fe/Pt(001) present the larger magnetization of the substrate and iron layer, the Kerr angles present significant variation at energy lower 10 eV only for Pt system roughly at 6.5 eV due to the variation of the off-diagonal in-plane conductivity in correspondence to this frequency. The lack of structure in Kerr signal for Pd and Pt system can be attributed - at least in part - to the fact that Pd and Pt elements the valence  $s$ -state is empty (while for the other material is filled by one electron). These results suggest that the hybridization effects between the Fe overlayer and the substrate and the relaxation of the Fe layer play an equally important role to determine the Kerr effect, and

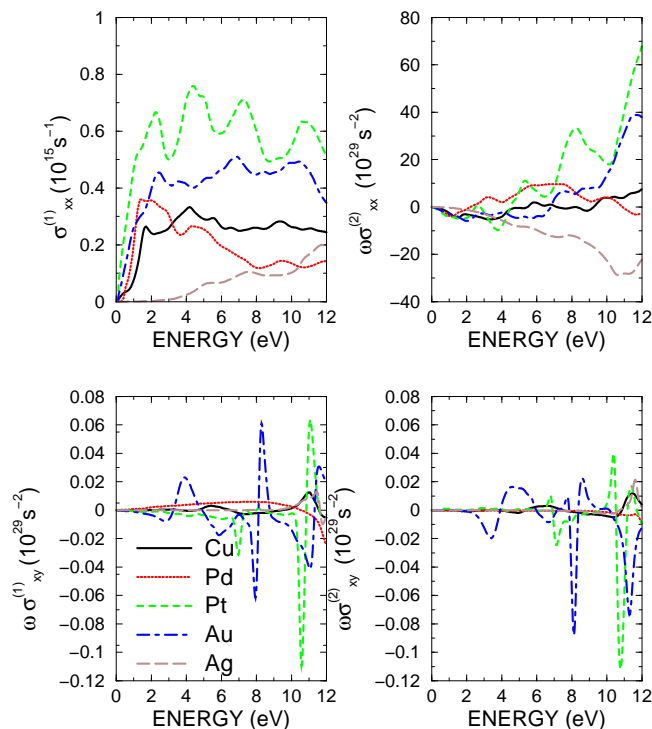


FIG. 3: Calculated optical conductivity for iron monolayer on different paramagnetic substrate: Cu (solid line), Pd (red line), Pt (green line), Au (blue line), and Ag (long dashed line). Top panel: diagonal absorptive optical conductivity (left), off-diagonal absorptive optical conductivity (right). Bottom panel: diagonal dispersive optical conductivity (left), off-diagonal dispersive optical conductivity (right).

only their combined effect can explain the observed Kerr spectra. For these reasons, a study as the one in Ref. 2 which treats only the change in the lattice parameters is insufficient to account for the differences in MOKE seen experimentally for Fe films on different substrates. To elaborate more on the conclusion of the last paragraph we discuss in detail our results displayed in Tab. I concerning the spin and orbital moments for an Fe monolayer on top of all five substrates. We consider firstly the three noble-metal substrates Cu, Au, and Ag, that belong to the same column of the periodic table. We remark firstly that the Fe spin moment depends mainly on the distance between the Fe layer and the substrate and thus it is comparable for the Ag and Au substrates, where also the in-plane Fe expansion is similar. The Fe orbital moment is comparable for both Cu and Au substrates, while for

the Ag substrate it is considerably larger. The orbital moment is not depending only on the spin moment of the Fe atom but also on the spin-orbit coupling of the substrate atoms as can be shown by perturbation theory.<sup>9</sup> The most important features are the magnetic properties of the first subsurface layer as they are more sensitive to hybridization effects than the Fe ones. Firstly we remark that the Fe atoms are ferromagnetically coupled to the substrate atoms. The Ag spin moment is considerably smaller than the Cu and Au ones suggesting that the hybridization between the Ag 4d and Fe 3d is considerably smaller than between the Cu 3d (Au 5d) and the Fe 3d electrons. This smaller hybridization is related to the absence of fine structures in the Kerr spectra of Fe/Ag(001) compared to the other two noble-metal systems. Finally the orbital moment at the Au site is considerably larger than for the other two noble-metal substrates due to the larger spin-orbit coupling of the 5d valence electrons compared to the 3d and 4d electrons in the case of Cu and Ag, respectively. The trends of the magnetization of Fe monolayer on Pd and Pt are opposite to those found for the noble metal since the magnetization of iron decrease for increasing lattice constant of the substrate. Pd and Pt have respectively one electron less than Ag and Au (in 5s or 6s shell). However the fine structure in the Kerr spectra is larger present in Pt but almost absent in Pd (at least up to 10 eV), similar trends are found in Ag and Au. This is a further proof of the role of hybridization of the *d*-shell as discussed above for the noble metals.

On summary, we have computed the complex Kerr angle for a monolayer of iron pseudomorphically grown on the [001] surface of Cu, Pd, Pt, Au, and Ag. Our results probe the importance of both the lattice relaxations and the hybridization effects between the Fe layer and the substrate on the calculation of the Kerr effect.

### Acknowledgment

The computer time was granted by IDRIS on the IBM RS6000 (Project No. 001266) and by Université Louis Pasteur of Strasbourg on the SGI Origin-2000 supercomputer. Financial support for this work has been provided by the TMR-Network of *Interface Magnetism* (contract ERBFMX96-0089) and the RT Network of *Computational Magnetoelectronics* (Contract RTN1-1999-00145) of the European Commission. This research was also supported in part by the National Science Foundation under Grant No. PHY99-07949.

<sup>1</sup> See, *e. g.*, H. Ebert, Rep. Prog. Phys. 59 (1996) 1665-1735, and references therein.

<sup>2</sup> A. Delin, O. Eriksson, B. Johansson, S. Auluck, and J. M. Wills, Phys. Rev. B 60 (1999) 14105-14114.

<sup>3</sup> J. Wills, O. Eriksson, M. Alouani, and D.L. Price, *Electronic Structure and Physical Properties of Solids: The Uses of the LMTO method*, Hugues Dreyssé (ed.), Lecture Notes on Physics-535, Springer (2000).

- <sup>4</sup> P. Hohenberg and W. Kohn, Phys. Rev. 136 (1964) B864-B871; W. Kohn and L. J. Sham, Phys. Rev. 140, (1965) A1133-A1138.
- <sup>5</sup> I. Galanakis, M. Alouani, and H. Dreyssé, Phys. Rev. B, 62 (2000) 3923-3928; I. Galanakis, A. Debernardi, M. Alouani, and H. Dreyssé, Surf. Sci. 482-5 (2001) 1030-1034.
- <sup>6</sup> A. Debernardi, I. Galanakis, M. Alouani, and H. Dreyssé, proc. EMRS-meeting 2001, Comp. Mater. Sci. **24**, 205-209 (2002).
- <sup>7</sup> Preliminary results concerning the magneto optical Kerr effect Cu and Ag were published in the proceeding of E-MRS meeting 2001 in Ref. 6. The present data are computed on a wider energy range.
- <sup>8</sup> T. Katayama, Y. Suzuki, Hiroyuki Awano, Y. Nishihara, and N. Koshizuka, Phys. Rev. Lett. 60 (1988) 1426-1429.
- <sup>9</sup> I. Galanakis, P. M. Oppeneer, P. Ravindran, L. Nordström, P. James, M. Alouani, H. Dreyssé, O. Eriksson, Phys. Rev. B 63 (2001) 172405-172408.